

## Claims:

1. A proton-conducting polymer membrane which comprises polyazoles and is coated with a catalyst layer and is obtainable by a process comprising the steps
  - A) preparation of a mixture comprising polyphosphoric acid,  
at least one polyazole (polymer A) and/or one or more compounds which are suitable for forming polyazoles under the action of heat according to step B),
  - B) heating of the mixture obtainable according to step A) under inert gas to temperatures of up to 400°C,
  - C) application of a layer using the mixture according to step A) and/or B) to a support,
  - D) treatment of the membrane formed in step C) until it is self-supporting,
  - E) application of at least one catalyst layer to the membrane formed in step C) and/or in step D).
2. The membrane as claimed in claim 1, characterized in that the support used in step C) has been provided with a catalyst-containing coating in order to provide the layer formed in step C) with a catalyst layer.
3. The membrane as claimed in claim 1 or 2, characterized in that the steps D) and E) are carried out simultaneously, with the membrane obtained in step C) being treated until it is self-supporting and provided with a catalyst layer in one step.
4. The membrane as claimed in one or more of the preceding claims, characterized in that the catalyst layer is applied by means of a powder process as in step E).
5. The membrane as claimed in one or more of the preceding claims, characterized in that the catalyst layer is applied in step E) by means of a process in which a catalyst suspension is used.

6. The membrane as claimed in claim 5, characterized in that the catalyst suspension comprises at least one organic, nonpolar solvent.
7. The membrane as claimed in claim 5 or 6, characterized in that the catalyst suspension comprises phosphoric acid and/or polyphosphoric acid.
8. The membrane as claimed in one or more of the preceding claims, characterized in that the catalyst layer is applied in step E) by means of a process in which a catalyst-containing coating is applied to a support and the catalyst-containing coating present on the support is subsequently transferred to the membrane obtained according to step C) and/or step D).
9. The membrane as claimed in claim 8, characterized in that the transfer of the catalyst-containing coating is effected by hot pressing.
10. The membrane as claimed in any of the preceding claims, characterized in that the mixture prepared in step A) comprises compounds which are suitable for forming polyazoles under the action of heat according to step B), with these compounds comprising one or more aromatic and/or heteroaromatic tetraamino compounds and one or more aromatic and/or heteroaromatic carboxylic acids or derivatives thereof which have at least two acid groups per carboxylic acid monomer, and/or one or more aromatic and/or heteroaromatic diamino-carboxylic acids.
11. The membrane as claimed in any of claims 1 to 9, characterized in that the mixture prepared in step A) comprises compounds which are suitable for forming polyazoles under the action of heat according to step B), with these compounds being obtainable by reaction of one or more aromatic and/or heteroaromatic tetraamino compounds with one or more aromatic and/or heteroaromatic carboxylic acids or derivatives thereof which have at least two acid groups per carboxylic acid monomer or of one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 400°C.

12. The membrane as claimed in claim 10 or 11, characterized in that aromatic and/or heteroaromatic tetraamino compounds used as compounds suitable for forming polyazoles comprise compounds selected from the group consisting of 3,3',4,4'-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine and 1,2,4,5-tetraamino-  
5 benzene.
13. The membrane as claimed in claim 10, 11 or 12, characterized in that aromatic and/or heteroaromatic carboxylic acids or derivatives thereof containing at least two acid groups per carboxylic acid monomer which are used as compounds suitable for forming polyazoles comprise compounds selected from the group consisting of isophthalic acid, terephthalic acid, phthalic acid, 5-  
10 hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid,  
15 2,5-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-  
20 naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, bis(4-carboxyphenyl) ether, benzophenone-4,4'-dicarboxylic acid, bis(4-carboxyphenyl) sulfone, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl)hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-  
25 carboxycinnamic acid, or their C1-C20-alkyl esters or C5-C12-aryl esters, or their acid anhydrides or their acid chlorides.
14. The membrane as claimed in claim 10, 11, 12 or 13, characterized in that the compounds suitable for forming polyazoles comprise aromatic tricarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides or tetracarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides.  
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15. The membrane as claimed in claim 14, characterized in that the aromatic tricarboxylic acids comprise compounds selected from the group consisting of 1,3,5-benzotricarboxylic acid (trimesic acid); 2,4,5-benzotricarboxylic acid (trimellitic acid); (2-carboxyphenyl)iminodiacetic acid, 3,5,3'-biphenyl-  
5 tricarboxylic acid; 3,5,4'-biphenyltricarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acid; naphthalene-1,4,5,8-tetracarboxylic acid, 3,5,3',5'-biphenyltetracarboxylic acid, benzophenonetetracarboxylic acid, 3,3',4,4'-biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxylic acid and 1,4,5,8-naphthalenetetracarboxylic  
10 acid.
16. The membrane as claimed in claim 14 or 15, characterized in that the content of tricarboxylic acids and/or tetracarboxylic acids is in the range from 0 to 30 mol%, preferably from 0.1 to 20 mol%, in particular from 0.5 to 10 mol%,  
15 based on dicarboxylic acid used.
17. The membrane as claimed in one of more of claims 10 to 16, characterized in that the compounds suitable for forming polyazoles comprise heteroaromatic dicarboxylic acids, tricarboxylic acids and/or tetracarboxylic acids containing at  
20 least one nitrogen, oxygen, sulfur or phosphorus atom in the aromatic.
18. The membrane as claimed in claim 17, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid,  
25 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazine-dicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid, and also their C1-C20-alkyl esters or C5-C12-aryl esters, or their acid anhydrides or their acid chlorides are used.
19. The membrane as claimed in claim 10 or 11, characterized in that the  
30 compounds suitable for forming polyazoles comprise diaminobenzoic acid and/or its monohydrochloride and dihydrochloride derivatives.

20. The membrane as claimed in one or more of the preceding claims, characterized in that the heating according to step B) is carried out after formation of a sheet-like structure according to step C).

5 21. The membrane as claimed in claim 1, characterized in that the treatment according to step D) is carried out at temperatures in the range from 0°C to 150°C in the presence of moisture.

10 22. The membrane as claimed in one or more of the preceding claims, characterized in that the treatment of the membrane in step D) is in the range from 10 seconds to 300 hours.

15 23. The membrane as claimed in one or more of the preceding claims, characterized in that the membrane formed after step D) and/or step E) is crosslinked by action of oxygen.

20 24. The membrane as claimed in one or more of the preceding claims, characterized in that a layer having a thickness of from 20 to 4000  $\mu\text{m}$  is produced in step C).

25 25. The membrane as claimed in one or more of the preceding claims, characterized in that the membrane formed after step D) has a thickness in the range from 15 to 3000  $\mu\text{m}$ .

26. The membrane as claimed in one or more of the preceding claims, characterized in that the catalyst layer has a thickness in the range from 0.1 to 50  $\mu\text{m}$ .

30 27. The membrane as claimed in one or more of the preceding claims, characterized in that the catalyst layer comprises catalytically active particles which have a size in the range from 0.1 to 10  $\mu\text{m}$ .

28. The membrane as claimed in one or more of the preceding claims, characterized in that the membrane provided with a catalyst layer comprises from 0.1 to 10 g/m<sup>2</sup> of a catalytically active substance.

5 29. The membrane as claimed in claim 27, characterized in that the catalytically active substance comprises particles comprising platinum, palladium, gold, rhodium, iridium und/or ruthenium.

10 30. The membrane as claimed in claim 28, characterized in that the catalytically active particles comprise carbon.

31. A membrane-electrode unit comprising at least one electrode and at least one membrane as claimed in one or more of claims 1 to 30.

15 32. A fuel cell comprising one or more membrane-electrode units as claimed in claim 31.